

**PATENT APPLICATION****IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Docket No: Q76524

Yutaka TOSAKI, et al.

Appln. No.: 10/616,916

Group Art Unit: 1713

Confirmation No.: 2409

Examiner: Kelechi Chidi Egwim

Filed: July 11, 2003

For: AQUEOUS DISPERSION TYPE PRESSURE-SENSITIVE ADHESIVE COMPOSITION AND  
PRESSURE-SENSITIVE ADHESIVE PRODUCT

**DECLARATION UNDER 37 C.F.R. § 1.132**

Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Yutaka Tosaki, hereby declare and state:

THAT I am a citizen of Japan;

THAT I have received a Bachelor's degree on March 1986 from Kobe University, Faculty  
of Engineering Department of Industrial Chemistry;

THAT I have been employed by Nito Denko Corporation since April of 1986, where I  
have been engaged in research and development relating to pressure-sensitive adhesive tapes  
to date.

I am a co-inventor of the invention described and claimed in the above-identified  
application.

I am familiar with the Office Action dated July 27, 2006. In regard to the rejection of  
present claims 1 and 3, the following experiments were carried out, either by me or under my  
direct supervision.

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The working examples of Iijima et al and Rosenski et al were reproduced as follows.

**Procedures for Reproducing the Working Examples in Iijima et al (U.S. 4,226,915)**

As the working example to be reproduced, I chose the formulation in line 13 of Table 1 in Example 1 of Iijima et al (the table below). The reason why this formulation was chosen is that poly (vinyl alcohol), which is shown as an example in the present application, is employed as a water-swellaable polymer in Iijima et al and because the amount of 8% by weight (10.4% by weight relative to 100% by weight of an acrylic copolymer) falls within the range of from 0.5 to 15 parts by weight per 100 parts of the acrylic pressure-sensitive adhesive composition as defined in present claim 1. I expect the other working examples in Iijima et al to basically show the same tendency as the working example reproduced herein.

A significant difference between the present invention and Iijima et al is that Iijima et al calls for a low molecular weight water-soluble polymer such as polyethylene glycol or glycerin with a molecular weight not exceeding 1000 as an essential component, whereas in the present invention, a specific polyalkylene glycol having a weight average molecular weight of from 100,000 to 3,000,000 alone is incorporated into a specific acrylic water dispersion type pressure-sensitive adhesive.

**Example 1 of Iijima et al**

| Material             | % by weight | % by weight | Manufacturer                    | Commercial product name |
|----------------------|-------------|-------------|---------------------------------|-------------------------|
| 2-ethyhexyl acrylate | 80          | 77          |                                 |                         |
| Methyl methacrylate  | 20          |             |                                 |                         |
| Polyvinyl alcohol    |             | 8           | Nippon Synthetic Chem. Ind. Co. | Gosenol GM-14           |
| Glycerin             |             | 15          |                                 |                         |

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According to the aforementioned working example of Iijima et al, a replication experiment was conducted using the following formulation. Since no description of the synthesis of the main component polymer is given except for the monomer composition ratios, the polymer employed in the replication experiment was synthesized by an ordinary monomer emulsion dropping method.

**Replication Experiment of Example 1 of Iijima et al (Replication Experiment ①)**

| Material                | % by weight | % by weight | Manufacturer                    | Commercial product name |
|-------------------------|-------------|-------------|---------------------------------|-------------------------|
| 2-ethylhexyl acrylate   | 80          | 77          | Toagosei Co.                    | 2EHA                    |
| Methyl methacrylate     | 20          |             | Mitsubishi Gas Chemical Co.     | MMA                     |
| Ammonium lauryl sulfate | 3           |             | Kao Co.                         | LATEMUL AD-25           |
| Polyvinyl alcohol       |             | 8           | Nippon Synthetic Chem. Ind. Co. | Gosenol GM-14           |
| Glycerin                |             | 15          | Wako Pure Chemical Co.          |                         |

To a reaction vessel equipped with a thermometer, a stirrer, a nitrogen introducing tube and a reflux cooling tube, 40 parts of water were charged, and nitrogen was introduced at room temperature for 1 hr. Thereafter, the content was heated to 70°C, and 0.4 parts of sodium persulfate dissolved in 10 parts of water were added. Then, a mixture obtained by emulsifying 160 parts of 2-ethylhexyl acrylate (a product of Toagosei Co., Ltd.) and 40 parts of methyl methacrylate (a product of Mitsubishi Gas Chemical Co., Inc.) into 69 parts of water with use of 25 parts of ammonium lauryl sulfate (LATEMUL AD-25, solid content of 24%, a product of Kao Corp.) was added dropwise over the period of 1 hr 40 min. During this addition, the temperature of the inner bath was kept at 70°C. Thereafter, by cooling the content and

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adjusting its pH to neutral by adding 2.4 parts of 10% aqueous ammonia at the inner bath temperature of 30°C, an emulsion of the main polymer was obtained.

To the solid content (77% by weight) of this emulsion, were added 8% by weight of poly(vinyl alcohol) (Gosenol GM-14; a product of Nippon Synthetic Chemical Co., Ltd.) and 15% by weight of glycerin (a product of Wako Pure Chemical Industries, Ltd.) as shown in the table above. The pressure-sensitive adhesive solution thus prepared was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to have a thickness of 20  $\mu$ m after drying, and dried to obtain a pressure-sensitive adhesive tape. (Replication Experiment ①).

Further, as amended, claim 1 recites a polyalkylene glycol. Under the circumstances, I also conducted comparative experiments using a polyalkylene glycol instead of the polyvinyl alcohol. As an example of the polyalkylene glycol, polyethylene glycol (Alkox E-30 manufactured by Meisei Kagaku Ind. co., Molecular weight of 300,000 to 500,000) was added in an amount of 1.54 part by weight based on 77 parts by weight of the polymers (corresponding to 2 parts by weight based on 100 parts by weight of the polymers). Further, I conducted a replication experiment in which glycerin was added in an amount of 15 parts by weight (Replication Experiment ②) and a replication experiment in which no glycerin was added (Replication Experiment ③). The conditions for producing a pressure-sensitive adhesive tape were the same as above.

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Replication experiment of Example 1 of Iijima et al. (Replication Experiment ②)

| Material                | % by weight | % by weight | Manufacturer                | Commercial product name |
|-------------------------|-------------|-------------|-----------------------------|-------------------------|
| 2-ethylhexyl acrylate   | 80          | 77          | Toagosei co.                | 2EHA                    |
| Methyl methacrylate     | 20          |             | Mitsubishi Gas Chemical co. | MMA                     |
| Ammonium lauryl sulfate | 3           |             | Kao co.                     | LATEMUL AD-25           |
| Polyethylene glycol     |             | 1.54        | Meisei Kagaku Ind. co.      | Alkox E-30              |
| Glycerin                |             | 15          | Wako Pure Chemical co.      |                         |

Replication experiment of Example 1 of Iijima et al. (Replication Experiment ③)

| Material                | % by weight | % by weight | Manufacturer                | Commercial product name |
|-------------------------|-------------|-------------|-----------------------------|-------------------------|
| 2-ethylhexyl acrylate   | 80          | 77          | Toagosei co.                | 2EHA                    |
| Methyl methacrylate     | 20          |             | Mitsubishi Gas Chemical co. | MMA                     |
| Ammonium lauryl sulfate | 3           |             | Kao co.                     | LATEMUL AD-25           |
| Polyethylene glycol     |             | 1.54        | Meisei Kagaku Ind. co.      | Alkox E-30              |
| Glycerin                |             | -           | Wako Pure Chemical co.      |                         |

**Replication Experiment for the Working Example in Rosenski et al (US 5,319,020)**

As the working example for the replication experiment, I chose Example 1 of Rosenski et al because it is the most typical example of the formulation of Rosenski et al. Meanwhile, I expect that the other working examples, in which various conditions are changed, including the kind of neutralizing agent, the molecular weight of PEG, the kind of acid, etc., to basically give results similar to those of Example 1.

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On this occasion, the replication experiment for Example 1 was carried out with the following formulation and procedure.

The results thus obtained clearly show the difference in the resulting product between the present invention (where the polyalkylene glycol is incorporated into the composition in the form of an aqueous solution after the polymerization of the acrylic polymer) and Rosenski et al (where a polyalkylene oxide plasticizer is incorporated during polymerization).

**Example 1 of Rosenski et al**

|    | Material                                       | Grams | Manufacturer      | Material Name and information on physical properties |
|----|--|-------|-------------------|--|
| A  | Deionized Water                                | 255   |                   |  |
|    | Sodium Acetate (buffer)                        | 0.675 |                   |  |
|    | PEG 8000 (plasticizer)                         | 90    | Union Carbide     | Polyethylene glycol (Mw8000)                         |
|    | Aerosol MA (anionic surfactant)                | 1.5   | American Cyanamid | Diethyl ester of sodium sulfosuccinic acid           |
|    | Abex 26S (anionic surfactant)                  | 1.35  | Rhone-Poulenc     | Alkyl phenol ether sulfate                           |
|    | Siponic L4 (nonionic surfactant)               | 3.0   | Rhone-Poulenc     | Polyoxyethylene (4) lauryl alcohol                   |
| B  | 2-Ethylhexyl Acrylate                          | 24    |                   |  |
|    | Vinyl Acetate                                  | 6     |                   |  |
| C  | Sodium Persulfate                              | 0.3   |                   |  |
|    | Deionized Water                                | 4.0   |                   |  |
| D1 | Deionized Water                                | 66    |                   |  |
|    | Abex 26S                                       | 13.8  |                   |  |
|    | Sodium Vinyl Sulfonate (stabilizing comonomer) | 3.0   |                   |  |
| D2 | 2-Ethylhexyl Acrylate                          | 205.5 |                   |  |
|    | Vinyl acetate                                  | 45.0  |                   |  |
|    | Monooctyl Maleate                              | 30.0  |                   |  |
|    | Acrylic Acid                                   | 1.5   |                   |  |
|    | Siponic L4                                     | 3.0   |                   |  |
| E  | Deionized Water                                | 24    |                   |  |
|    | Sodium Persulfate (initiator)                  | 0.75  |                   |  |
|    | Ammonium Hydroxide                             | 9.9   |                   |  |

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|   | Material                              | Grams | Manufacturer | Material Name and information on physical properties |
|---|---------------------------------------|-------|--------------|--|
|   | (neutralizing agent)                  |       |              |  |
| F | t-Butyl Hydroperoxide (scavenger)     | 0.3   |              |  |
|   | Deionized Water                       | 1.0   |              |  |
| G | Sodium Metabisulfite (reducing agent) | 0.6   |              |  |
|   | Deionized Water                       | 4.5   |              |  |

Based on the above-described working example, the replication experiment was carried out with the following formulation.

**Replication Experiment for Example 1 of Rosenski et al**

|    | Material                                      | Grams | Manufacturer           | Material Name and information on physical properties |
|----|---|-------|------------------------|--|
| A  | Deionized Water                               | 253   |                        |  |
|    | Sodium Acetate (buffer)                       | 0.675 | Kishida Chemical Co.   |  |
|    | PEG 6000 (plasticizer)                        | 90    | Kishida Chemical Co.   | Polyethylene glycol (Mw8000)                         |
|    | Sodium dialkyl sulfosuccinate                 | 2.143 | Kao Co.                | PELEX OTP 70%  |
|    | Sodium alkyl diphenyl ether disulfonate       | 2.7   | Kao Co.                | PELEX SS-H 50%                                       |
|    | Polyoxyethylene lauryl ether                  | 3.0   | Kao Co.                | EMULGEN 104P, 100%                                   |
| B  | 2-Ethylhexyl acrylate                         | 24    | Toagosei Co.           |  |
|    | Vinyl Acetate                                 | 6     | Denki Kagaku Kogyo Co. |  |
| C  | Sodium Persulfate                             | 0.3   | Kishida Chemical Co.   |  |
|    | Deionized Water                               | 4.0   |                        |  |
| D1 | Deionized Water                               | 52.2  |                        |  |
|    | Sodium alkyl diphenyl ether disulfonate       | 27.6  | Kao Co.                | PELEX SS-H, 50%                                      |
|    | Sodium allylsulfonate (stabilizing comonomer) | 3.0   | Tokyo Kasei Kogyo Co.  |  |
| D2 | 2-Ethylhexyl Acrylate                         | 205.5 | Toagosei Co.           |  |
|    | Vinyl acetate                                 | 45.0  | Denki Kagaku Kogyo Co. |  |
|    | Diethyl Maleate                               | 30.0  | Wako Pure Chemical Co. |  |

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|   | Material                                | Grams | Manufacturer         | Material Name and information on physical properties |
|---|---|-------|----------------------|--|
|   | Acrylic Acid                            | 1.5   | Toagosei Co.         |  |
|   | Polyoxyethylene lauryl ether            | 3.0   | Kao Co.              | EMULGEN 104P, 100%                                   |
| E | Deionized Water                         | 24    |                      |  |
|   | Sodium Persulfate (initiator)           | 0.75  | Kishida Chemical Co. |  |
|   | Ammonium Hydroxide (neutralizing agent) | 9.9   | Kishida Chemical Co. |  |
| F | t-Butyl Hydroperoxide (scavenger)       | 0.429 | Kayaku Akzo Co.      | KayabutylH70, 70%                                    |
|   | Deionized Water                         | 0.871 |                      |  |
| G | Sodium Metabisulfite (reducing agent)   | 0.6   | Kishida Chemical Co. |  |
|   | Deionized Water                         | 4.5   |                      |  |

**Remarks Regarding the materials used in Example 1:**

- Since no description is given on the length of the alkyl group for "Abex 26s" among the surfactants, a typical commercial product was chosen.
- Since sodium vinylsulfonate is not easily available, sodium allylsulfonate was used in its place.
- Since mono-octyl maleate is not easily available, dioctyl maleate was used in its place.
- With regard to the remaining materials, those having the same chemical formulae were used, which for convenience, were obtained from manufacturers easily accessible in Japan.
- Some quantity differences in the formulation are the result of re-calculation so that accurate solid contents were employed. This is because the surfactants, etc., are in aqueous solution form.

The polymerization formulation was faithfully reproduced according to Example 1.

The pressure-sensitive adhesive solution was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to give a thickness of 20  $\mu$ m after drying, and dried to give a pressure-sensitive adhesive tape.



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### Results of the Replication Experiments

The results of measuring the wet-surface adhesive forces of the above-described two kinds of pressure-sensitive adhesive tapes by the method as set forth in the working example of the present application are shown in Table 1 below.

**TABLE 1**

|  | Wet surface adhesive force (N/18 mm) |
|--|--------------------------------------|
| Replication experiment ① for formulation in line 13 of Example 1 in Iijima et al. (with glycerin)    | 0.02                                 |
| Replication experiment ② for formulation in line 13 of Example 1 in Iijima et al. (with glycerin)    | 0.04                                 |
| Replication experiment ③ for formulation in line 13 of Example 1 in Iijima et al. (without glycerin) | 0.33                                 |
| Example 1 of Rosenski et al.   | Cohesive failure                     |

As a result of replicating the working example of Iijima et al (Replication Experiment ①), the wet-surface adhesive force proved to be 0.02 N/18 mm. I attribute such a low value to the incorporation of glycerin having a low molecular weight. Namely, when a low molecular weight hydrophilic ingredient such as glycerin is formulated in a pressure-sensitive adhesive, it is naturally present at the surface, thus enhancing the hydrophilicity of the surface of the pressure-sensitive adhesive. Thus, when the tape is attached to a wet surface of an adherend, energetically stable moisture is present between the adherend and the surface of the pressure-sensitive adhesive. As such, a low wet-surface adhesive force results due to the moisture that is not absorbed into the interior of the pressure-sensitive adhesive. Accordingly, differing from Iijima, the present claims define a lower limit of the molecular weight of the hydrophilic polymer

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of 100,000. By setting the lower limit of the molecular weight at 100,000, easy migration of the hydrophilic polymer to the surface is suppressed.

Further, in the system in which polyethylene glycol was added instead of polyvinyl alcohol, when glycerin was added (Replication Experiment ②), the wet-surface adhesive force proved to be 0.04 N/18 mm. In the same system, when no glycerin was added (Replication Experiment ③), the wet-surface adhesive force proved to be 0.33 N/18 mm. These values are quite different from each other depending upon the presence or absence of glycerin. It is understood from the results that the adhesive force to the wet surface is reduced by adding glycerin having the low molecular weight.

On the other hand, the replication experiment for the working example of Rosenski et al resulted in cohesive destruction, and an ordinary adhesive force could not be measured. I attribute these results to the low molecular weight of the polymer obtained by polymerization, whereby the cohesive force was low. It seems that the low molecular weight is caused by a decrease in polymerization speed due to (1) the viscosity increase in the system caused by the presence of the polyalkylene oxide plasticizer as a hydrophilic polymer during polymerization and by the protection of the particle surface with the hydrophilic component, and (2) the polymerization under a substantially neutral condition, not acid condition.

In the present invention, apart from Rosenski et al, the hydrophilic polymer is added after polymerization, and this difference also promotes good wet-surface adhesive force.

From these results, it is evident that a distinct difference between the present invention and Iijima et al is that Iijima et al essentially requires the addition of a low molecular weight water-soluble polymer, whereas the present invention sets the lower limit of an average

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molecular weight of 100,000. As shown above, the low molecular weight water-soluble polymer has an adverse effect on initial adhesive force to a dewy or wet surface, contrary to the object of the present invention. In the present invention, the polyalkylene glycol has a weight average molecular weight of from 20,000 to 5,000,000 and more preferably from 100,000 to 3,000,000 (as presently claimed). Page 16, lines 20-22. As further taught therein, "when the molecular weight is lower than 20,000, a drawback occurs that the effect improving the adhesive force to a dewy or wet surface deteriorates". Thus, the disadvantage of a low molecular weight water-soluble polymer as taught by Iijima et al has been verified. Accordingly, in disclosing a formulation for a pressure-sensitive adhesive of rather deteriorated performance, Iijima et al has no relevance in achieving the object of the present invention.

Further, a clear difference between the present invention and Rosenski et al is that, in contrast to Rosenski et al where polymerization is conducted in the presence of a polyalkylene oxide, the present invention adds a polyalkylene oxide in the form of an aqueous solution after polymerization. This difference in the mode of addition affects the polymerization reactivity, thus resulting in the failure of Rosenski et al to exhibit good initial adhesive force to a dewy surface.

I confirmed that, while the polyalkylene oxide in Rosenski et al is adsorbed on the particle surface since it is added prior to polymerization, in the present invention, where a polyalkylene oxide is added after polymerization, the compound, according to electron micrography, is in an aggregated state dispersed in the pressure-sensitive adhesive (not adsorbed on particle surface) after coating and drying of the pressure-sensitive adhesive composition. Furthermore, in the present invention, the dispersed aggregated state exhibits a

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high initial adhesive force to a dewy or wet surface, which is a main object of the present invention.

For example, in emulsion polymerization for the synthesis of an emulsion-type pressure-sensitive adhesive in general, it is well known that, even if the final composition is the same, different particle size, particle size distribution and the molecular weight of the polymer are obtained depending on whether the entire amount of a monomer emulsion is added to the polymerization system at once, or gradually drop by drop.

Once such variation of physical properties is introduced to the same composition, the properties of the resulting pressure-sensitive adhesive tape are also expected to vary.

Since the present invention differs from Rosenski et al in objective as well as technical content, in my opinion Rosenski et al does not lead one of ordinary skill to the present invention in any sense.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: July 13, 2006

Yutaka Tosaki  
Name of Declarant